



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : H01J 01/30, C23C 16/00	A1	(11) International Publication Number: WO 00/30141 (43) International Publication Date: 25 May 2000 (25.05.00)
(21) International Application Number: PCT/US99/26232 (22) International Filing Date: 4 November 1999 (04.11.99) (30) Priority Data: 09/191,728 12 November 1998 (12.11.98) US (71) Applicant: THE BOARD OF TRUSTEES OF THE LELAND STANFORD JUNIOR UNIVERSITY [US/US]; 900 Welch Road, Suite 350, Palo Alto, CA 94304-1850 (US). (72) Inventors: DAI, Hongjie; 365 Pescadero Tr., Sunnyvale, CA 94507 (US). FAN, Shoushan; Building SW5-1-402, Tsinghua University, Beijing (CN). CHAPLINE, Michael; 2504 Round Hill Drive, Alamo, CA 94507 (US). FRANKLIN, Nathan; 350 Sharon Park Drive, #H-205, Menlo Park, CA 94025 (US). TOMBLER, Thomas, Jr.; 121 Campus Drive, Apartment 1409A, Stanford, CA 94305 (US). (74) Agent: ALBOSZTA, Marek; 426 Lowell Avenue, Palo Alto, CA 94301 (US).		(81) Designated States: CA, JP, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published With international search report.
(54) Title: SELF-ORIENTED BUNDLES OF CARBON NANOTUBES AND METHOD OF MAKING SAME <div data-bbox="518 1140 1071 1575" data-label="Image"> </div>		
(57) Abstract <p>A field emission device (20) having bundles (28) of aligned parallel carbon nanotubes on a substrate (22). The carbon nanotubes (28) are oriented perpendicular to the substrate (22). The carbon nanotube bundles (28) may be up to 300 microns tall, for example. The bundles (28) of carbon nanotubes extend only from regions of the substrate (22) patterned with a catalyst material (26). Preferably, the catalyst material (26) is iron oxide. The substrate (22) is preferably porous silicon, as this produces the highest quality, most well-aligned nanotubes. Smooth, nonporous silicon or quartz can also be used as the substrate. The method of the invention starts with forming a porous layer on a silicon substrate by electrochemical etching. Then, a thin layer of iron is deposited on the porous layer in patterned regions. The iron is then oxidized into iron oxide, and then the substrate is exposed to ethylene gas at elevated temperature. The iron oxide catalyzes the formation of bundles of nanotubes. The iron oxide catalyzes the formation of bundles of aligned parallel carbon nanotubes which grow perpendicular to the substrate surface. The height of the nanotube bundles above the substrate is determined by the duration of the catalysis step. The nanotube bundles only grow from the patterned regions.</p>		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece			TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	NZ	New Zealand		
CM	Cameroon	KR	Republic of Korea	PL	Poland		
CN	China	KZ	Kazakhstan	PT	Portugal		
CU	Cuba	LC	Saint Lucia	RO	Romania		
CZ	Czech Republic	LI	Liechtenstein	RU	Russian Federation		
DE	Germany	LK	Sri Lanka	SD	Sudan		
DK	Denmark	LR	Liberia	SE	Sweden		
EE	Estonia			SG	Singapore		

PCT Patent Application entitled

10

**Self-Oriented Bundles of Carbon Nanotubes and Method of Making
Same**

15

FIELD OF THE INVENTION

This invention relates generally to carbon nanotubes. The present invention also relates to methods for making bundles of aligned
20 carbon nanotubes. The present invention is also related to carbon nanotube field emission devices such as used in flat panel displays.

BACKGROUND

25 Field emission devices have potential applications in flat panel displays. Field emitters used in flat panel displays must be stable, long lasting and should have a relatively uniform emission over the surface of the display.

30 Carbon nanotubes are very small tube-shaped molecules having the structure of a graphite molecule rolled into a tube. Carbon nanotubes are electrically conductive along their length, chemically stable, and can have very small diameters (much less than 100 nanometers) and large aspect ratios (length/diameter).

35 Due to these properties and other properties, it has been suggested that carbon nanotubes can be used as field emission devices.

However, it has been unclear how to realize a field emission
40 device exploiting carbon nanotubes. They are difficult to work with in bulk, and, on a microscopic level, often form an impossibly tangled mess resembling a hairball. To produce a useful field emission device for flat panel displays, the carbon

- 5 nanotubes should be patterned into individual field emitters. A problem with present methods of making carbon nanotube field emitters is that it is not clear how to pattern the carbon nanotube to provide arrays of emitters.
- 10 US Patent no. 5,773,921 to Keesman et al. discloses a field emission device which employs sharp-edged graphite wafers. Carbon nanotubes are disposed on the sharp edge of the graphite wafers and help to increase field emission. The carbon nanotubes are disposed on the graphite wafers by sputtering a nearby graphite
- 15 target. The carbon nanotubes are not aligned in any way.

- US Patent no. 4,272,699 to Faubel et al. discloses an ion source which uses carbon fiber field emitters. The carbon fibers are bundled together and an electric field is applied to the bundle.
- 20 The carbon fibers are held by a macroscopic mechanical device that holds the fibers parallel. Such a mechanical device cannot be used with carbon nanotubes, which are orders of magnitude smaller than the carbon fibers used by Faubel et al.
- 25 US Patent no. 5,773,834 to Yamamoto et al. describes a method of making carbon nanotubes on the surface of a carbon-containing substrate by ion bombardment. The carbon nanotubes produced can be used as field emitters. The carbon nanotubes produced according to Yamamoto are not aligned, and in particular, are not
- 30 aligned perpendicular to the substrate. Also, Yamamoto does not disclose how to pattern the substrate to provide individual field emitters.

- Yet another problem with present methods for making carbon
- 35 nanotube field emitters is that scale-up to large wafers is difficult or not possible.

- There exists a need in the art for a method of producing aligned carbon nanotubes. In particular, such aligned carbon nanotubes
- 40 can be used as superior field emission devices.

5

OBJECTS AND ADVANTAGES

Accordingly, it is a primary object of the present invention to provide:

10

1) a method of making aligned bundles of carbon nanotubes; and

2) a field emission device using aligned bundles of carbon nanotubes as field emitters;

15

3) a field emission device that can have arrays of emitters;

4) a method of making field emitters that can be scaled to large substrates.

20

These and other objects and advantages will be apparent upon reading the following description and accompanying drawings.

SUMMARY

25

These objects and advantages are attained by a field emission device having a refractory substrate composed of silicon or quartz, a catalyst material on top of the substrate, and a bundle of aligned parallel carbon nanotubes extending from the catalyst material in a direction perpendicular to the substrate.

30

Preferably, the substrate has a top layer of porous silicon, with the catalyst material disposed on the porous layer. Particularly, the porous layer can be composed of an upper nanoporous layer with small pores on top of a macroscopic porous layer with larger pores. The catalyst material is preferably iron oxide.

35

The substrate may also have a smooth, nonporous surface, or a rough surface.

40

The carbon nanotube bundles may be within 10-22 nanometers in diameter and may be up to 300 microns tall. Also, the carbon nanotubes may be multi-walled.

Preferably, the catalyst material is confined to a patterned region. This results in the bundle extending from the patterned

5 region. The bundle has the same footprint size and shape as the patterned region of the catalyst material.

The carbon nanotube bundles may have a flat top, or a bowl-shaped top.

10

The present invention also includes a method of making bundles of aligned carbon nanotubes on a substrate of silicon or quartz. The method includes the steps of depositing a catalyst material on a top surface of the substrate, and then exposing the substrate to a

15

Preferably, the substrate is a silicon substrate with a porous top surface. The top surface may be made porous by electrochemical etching.

20

The catalyst material is preferably iron oxide. The iron oxide may be deposited by depositing a thin film of iron, and then oxidizing the iron film. The iron film may be oxidized by exposing it to oxygen at elevated temperature. Preferably, the

25

The carbon containing gas may be ethylene.

DESCRIPTION OF THE FIGURES

30 Fig. 1 shows a field emission device according to the present invention.

Fig. 2 shows a graph of field emission characteristics of a field emission device according to the present invention.

35 Fig. 3 illustrates a method according to the present invention for making bundles of aligned carbon nanotubes on a porous silicon substrate.

Fig. 4 illustrates a methods according to the present invention for making bundles of aligned carbon nanotubes on a smooth, nonporous silicon substrate.

40 Fig. 5 shows a field emission device with many nanotube bundles. Each bundle can provide field emission to illuminate a single pixel in a flat panel display.

5

DETAILED DESCRIPTION

Fig. 1 shows a field emission device 20 according to a preferred embodiment of the present invention. The device has a substrate 22 with a porous top layer 24. The substrate 22 and top layer 24 are made of silicon, although other substrate materials can also be used. Disposed on the porous top layer 24 are patterns of catalyst material 26. Preferably, the catalyst material 26 is a thin film of iron oxide. Extending from the catalyst material patterns 26 are carbon nanotube bundles 28, which are perpendicular to the substrate 22. The carbon nanotubes comprising the bundles are parallel and perpendicular to the substrate 22. The carbon nanotubes comprising the bundles are typically about 10-22 nanometers in diameter.

The bundles 28 may be about 10-250 microns wide 30, and up to or exceeding 300 microns in height 32. The nanotube bundles have the same width 30 as the catalyst material patterns 26. More generally, the bundles 28 have the same 'footprint' size and shape as the catalyst material patterns 26. The nanotube bundles 28 can have a flat top 34, or can have a bowl-shaped top 36, depending on the process parameters used to make the nanotube bundles. There are no individual stray nanotubes extending away from the blocks.

The nanotube bundles 28 have sharp edges 38 and corners which serve as field emission regions. Since the substrate can be conductive (e.g. doped silicon) and the nanotubes are conductive along their length (parallel with height 32), electrical connections to the bundles 28 are made simply by connecting to the substrate 22.

35

Fig. 2 shows field emission characteristics for four nanotube bundles on a n^+ type silicon substrate. The bundles each have a footprint of 250 microns X 250 microns and are 160 microns tall. The anode is an aluminum coated silicon wafer 200 microns above the substrate. The data is taken in a vacuum chamber at 3×10^{-7} Torr base pressure. Note that the electric field is computed by using the applied voltage divided by the distance from the tops of the bundles to the anode (40 microns). The current density

5 reaches 10mA/cm² at an electric field strength of 5 volts/micron. After continuous emission for a week at 2mA/cm² the bundles show no indication of damage under a scanning electron microscope.

The present invention includes methods of making the carbon
10 nanotube bundles attached to substrates. Fig. 3 illustrates a preferred method for making the nanotube bundles. First, in step A, a highly P-doped n⁺ type silicon substrate 22 (100 top surface, resistivity 0.008-0.018 Ohm-cm) is electrochemically etched in 1:1 HF (49% in water) ethanol with an anodization current density of
15 10mA/cm² (typical etching time is 5 minutes). This forms a thin nanoporous layer 42 (pore size~3 nanometers) on top of a macroporous layer 44 (pore size~100 nanometers). Layers 42, 44 are generally referred to as a porous layer 24 in Fig. 1. Next, in step B, the porous layer 42 is patterned with a 5 nanometer
20 thick iron film 46 by e-beam evaporation through a shadowmask 48. After deposition of iron, the substrate is annealed in air at 300°C overnight. This annealing step oxidizes the surface of the silicon as well as the iron, converting the iron patterns 46 into catalytically active iron oxide patterns 26. The resulting
25 silicon dioxide layer formed on the porous silicon prevents the porous structure of layers 42, 44 from collapsing during the following high temperature chemical vapor deposition (CVD) step. Next, in step C, the substrate 22 is placed in a 2-inch tube reactor 49 housed in a tube furnace. The furnace is heated to
30 700°C in flowing argon. Then, at 700°C, the argon supply is turned off, and ethylene 50 is flown through the tube reactor at a rate of 1000 sccm/min for 15-60 minutes. The boat for the substrates is sealed at one end, and the sealed end is placed downstream in the furnace. While ethylene 50 is flowing, the iron
35 oxide patterns 26 catalyze the growth of carbon nanotube bundles 28 which grow perpendicular to the substrate 22. The width of the bundles 28 is the same as the width of the iron oxide patterns 26.

The above method for making nanotube bundles produces mainly
40 flat-top bundles, although sometimes bowl-shaped bundles are also produced. Bowl-shaped bundles are produced when nanotubes in the middle of a bundle grow slower than the nanotubes at the outer edges of a bundle.

5

The height of the bundles 28 is determined by the duration that the substrates are exposed to ethylene at high temperature. In a particular experiment conducted by applicants, reaction times of 5, 15, 30, and 60 minutes produced bundles 31, 98, 163, and 240
10 microns tall, respectively. The growth rate is observed to be linear initially and then falls off at longer reaction times. As the aspect ratio (height:width) of the bundles approaches 5:1, some bundles may become tilted, but refuse to fall down.

15 The bundles can have footprint sizes as small as 2 microns on a side and be 15 microns tall, and still remain standing on the substrate.

Since the carbon nanotubes grow only from those regions that have deposited iron oxide, patterning the iron oxide results in carbon
20 nanotubes growing only from those regions with iron oxide. This provides accurate control of the size, shape, and distribution of the bundles on the substrate surface. As a particular advantage, patterning of the catalyst provides the ability to fabricate
25 arrays of individual field emitters. Each bundle can provide field emission for a single pixel in a flat panel display. Each bundle can be individually controlled by connecting patterned metallization lines to the bundles. Further, the arrays can be formed on large substrates. The size of the substrate is only
30 limited by the size of the tube furnace. The substrate can be several inches across, for example. Many field emitter arrays have been fabricated on 3cm X 3cm silicon substrates.

The method of the present invention produces carbon nanotube
35 bundles having a high density. The nanotubes are held together by Van der Waals interactions. The nanotubes grown according to the present method are in the size range of about 16 ± 6 nanometers and are aligned parallel in a direction perpendicular to the substrate
22. The carbon nanotubes are aligned perpendicular to the
40 substrate regardless of the orientation of the substrate in the furnace. The carbon nanotubes are generally multi-walled.

5 The porous layer acts as an excellent catalyst support. During the 300°C annealing step, iron oxide particles form with a narrow size distribution due to their strong interactions with the porous layer. The strong interactions also prevent the iron oxide particles from sintering at elevated temperatures.

10

It has been confirmed by the applicants that the nanotube bundles grow in 'base growth' mode. This has been established by physically removing nanotube bundles from the substrate and observing that the substrates remain capable of growing carbon
15 nanotubes (i.e. the iron oxide catalyst patterns remain on the substrate).

20

The present method of growing aligned nanotubes is very different from prior art approaches of growing aligned nanotubes. In prior art approaches, carbon nanotube alignment is provided by confining the growth of nanotubes to channels in porous silica or the channels of alumina membranes. In the present method, the carbon nanotubes are self aligned, without being confined to a channel, pore, or hole. The carbon nanotubes of the present method
25 spontaneously align themselves in free space.

30

It is important to note that the holes and pores in the porous layers 42, 44 are not aligned or oriented in any way. For example, they are not necessarily aligned perpendicular to the substrate 22. The holes and pores of the porous layer 24
generally have random orientations.

35

The present invention also includes a method of producing aligned carbon nanotubes on a nonporous smooth silicon substrate. The present method for growing aligned carbon nanotubes on nonporous silicon substrates is largely the same as the method for porous substrates. **Fig. 4** illustrates a first step in the present method for producing aligned carbon nanotube bundles on a smooth nonporous silicon substrate 40. A thin film (e.g. 5 nanometers
40 thick) of iron 46 is deposited through a shadowmask 48 using the same technique as in step B of **Fig. 3**. The nonporous substrate 40 has a native oxide layer which is left intact prior to the iron deposition. The 300°C annealing and 700°C CVD nanotube growth

5 steps for nonporous substrates 40 are the same as in the method described above for porous substrates. The CVD process forms carbon nanotube bundles which extend from iron oxide patterns perpendicularly to the nonporous substrate 40.

10 The overall features of carbon nanotubes grown on nonporous silicon are similar to the features of nanotubes grown on porous silicon. However, in contrast to porous silicon substrates, nanotube bundles with aspect ratios greater than 5 tend to fall onto the substrate 40. Nanotube bundles grown on smooth nonporous

15 substrates are not as strongly bound to the substrate. Also, carbon nanotube bundles grown on nonporous silicon substrates tend to have higher defect densities, and tend to be less well aligned than bundles grown on porous silicon. Further, carbon nanotubes grow about 50% faster on porous silicon compared to nonporous

20 silicon. For these reasons, the use of porous silicon substrates is highly preferred.

Fig. 5 shows a field emission device that can be used in a flat panel display. The device has many carbon nanotube bundles 28.

25 Each bundle 28 can provide field emission for a single pixel in a flat panel display.

It is also noted that the iron catalyst does not necessarily need to be deposited using physical vapor deposition techniques,

30 although this is preferred. For example, catalyst materials can be deposited as iron salts dissolved in a carrier solvent. The solvent is then deposited on the substrate and allowed to dry, leaving the iron salt. The iron salt may then need to be 'activated' by exposing it to high temperature so that it

35 decomposes into active catalyst material.

It is also noted that the substrate does not necessarily need to be silicon, although silicon, and particularly, porous silicon, is preferred. The substrate can also be quartz. In the present

40 application, silicon, porous silicon, and quartz are understood to be refractory materials.

5 The present method for making bundles of aligned carbon nanotubes
may also work on substrates such as ceramics, alumina, sapphire,
and silica, for example. The substrate must be able to tolerate
the high temperatures (about 700°C) used in the CVD process
without melting or disintegrating. For best results, the substrate
10 should have a rough and complex surface topology.

It is also noted that the substrate can have a rough texture which
is neither smooth nor 'porous'. Generally, however, extremely
complex substrate surface topologies are preferred because they
15 produce fast growing nanotubes with few defects that are strongly
bound to the substrate.

It will be clear to one skilled in the art that the above
embodiment may be altered in many ways without departing from the
scope of the invention. Accordingly, the scope of the invention
20 should be determined by the following claims and their legal
equivalents.

5

CLAIMS

What is claimed is:

1. A field emission device comprising:
 - 10 a) a substrate composed of refractory material;
 - b) a catalyst material deposited on a top surface of the substrate;
 - 15 c) a bundle of parallel carbon nanotubes extending from the catalyst material in a direction perpendicular to the substrate.
2. The field emission device of claim 1 wherein the
20 substrate is a silicon substrate.
3. The field emission device of claim 2 wherein the silicon substrate has a porous top layer and the catalyst material is disposed on the porous top
25 layer.
4. The field emission device of claim 3 wherein the porous top layer comprises a first porous layer on top of a second porous layer, and wherein the
30 pores of the first layer are smaller than the pores of the second layer.
5. The field emission device of claim 1 wherein the catalyst material comprises iron oxide.
- 35 6. The field emission device of claim 1 wherein the silicon substrate has a smooth top surface.
7. The field emission device of claim 1 wherein the
40 silicon substrate has a rough top surface.

- 5 8. The field emission device of claim 1 wherein the
 carbon nanotubes have diameters in the range of 10-22
 nanometers.
- 10 9. The field emission device of claim 1 wherein the
 carbon nanotubes are multi-walled.
10. The field emission device of claim 1 wherein the
 carbon nanotube bundles are less than 300 microns
 tall.
- 15 11. The field emission device of claim 1 wherein the
 catalyst material is confined to a patterned region.
12. The field emission device of claim 11 wherein the
20 carbon nanotube bundle has the same footprint
 size and shape as the patterned region.
13. The field emission device of claim 1 wherein the
 carbon nanotube bundle has a flat top.
- 25 14. The field emission device of claim 1 wherein the
 carbon nanotube bundle has a bowl-shaped top.
- 30 15. A method of making bundles of aligned carbon nanotubes on a
 top surface of a refractory substrate, the method comprising
 the steps of:
- a) depositing a catalyst material on the top surface of
 the refractory substrate;
- 35 b) exposing the catalyst material to a carbon containing
 gas at elevated temperature.
- 40 16. The field emission device of claim 15 wherein the
 substrate is a silicon substrate.

- 5 17. The method of claim 16 further comprising the
step of forming a porous top layer on the silicon
substrate before step (a).
- 10 18. The method of claim 17 wherein the porous top
layer is produced by electrochemical etching.
- 15 19. The method of claim 15 wherein the catalyst material
comprises iron oxide particles.
- 20 20. The method of claim 15 wherein step (a) comprises the
steps of depositing an iron film and then oxidizing
the iron film.
- 25 21. The method of claim 20 wherein oxidation of the
iron film is performed by exposing the iron film
to oxygen at elevated temperature.
22. The method of claim 20 wherein deposition of the
iron film is performed by physical vapor
deposition.
- 30 23. The method of claim 20 wherein the iron film is 5
nanometers thick.
24. The method of claim 15 wherein the carbon containing
gas is ethylene.

1/3

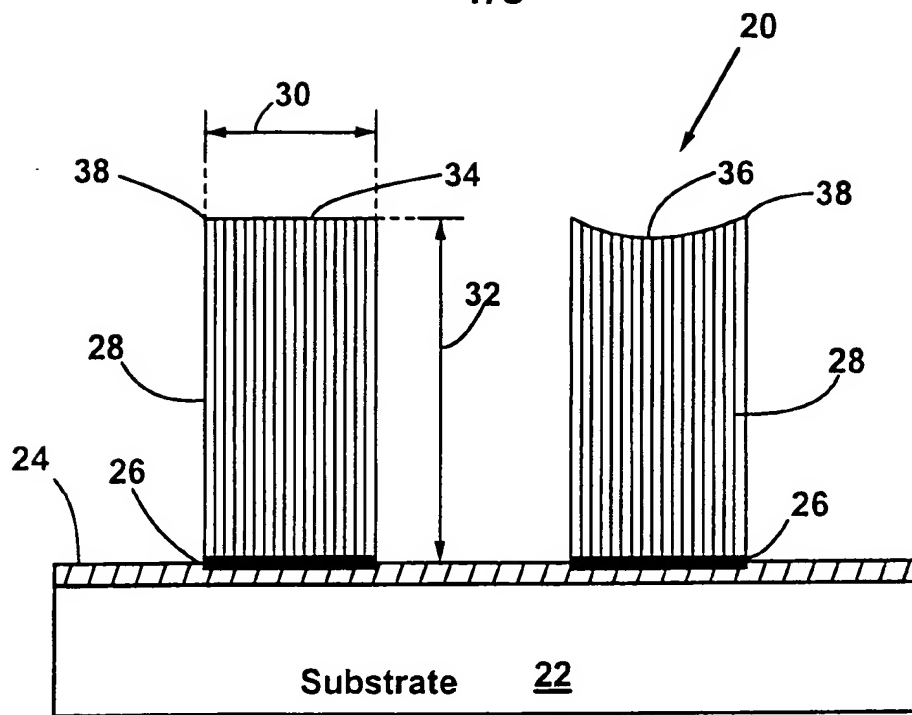


Fig. 1

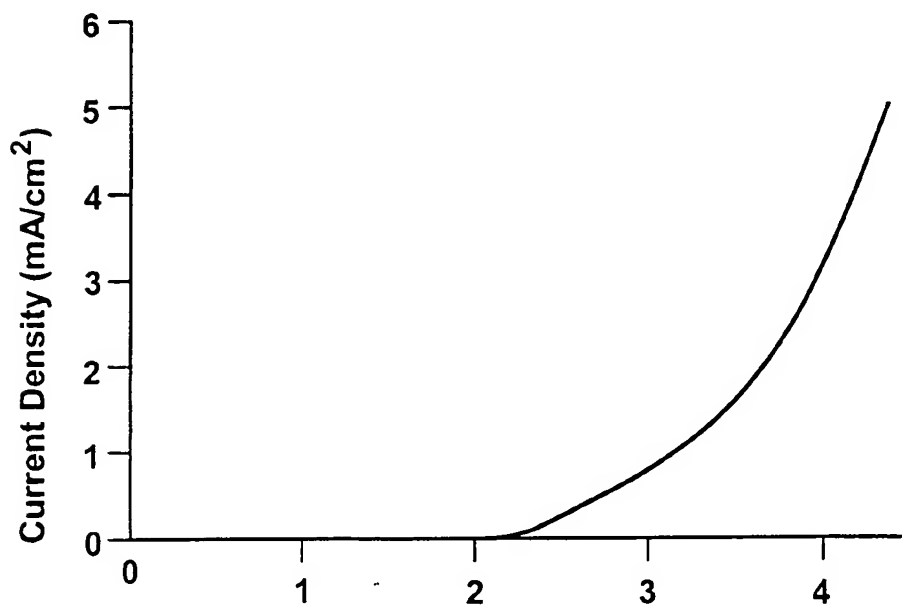


Fig. 2

Electric Field (Volts/micron)

2/3

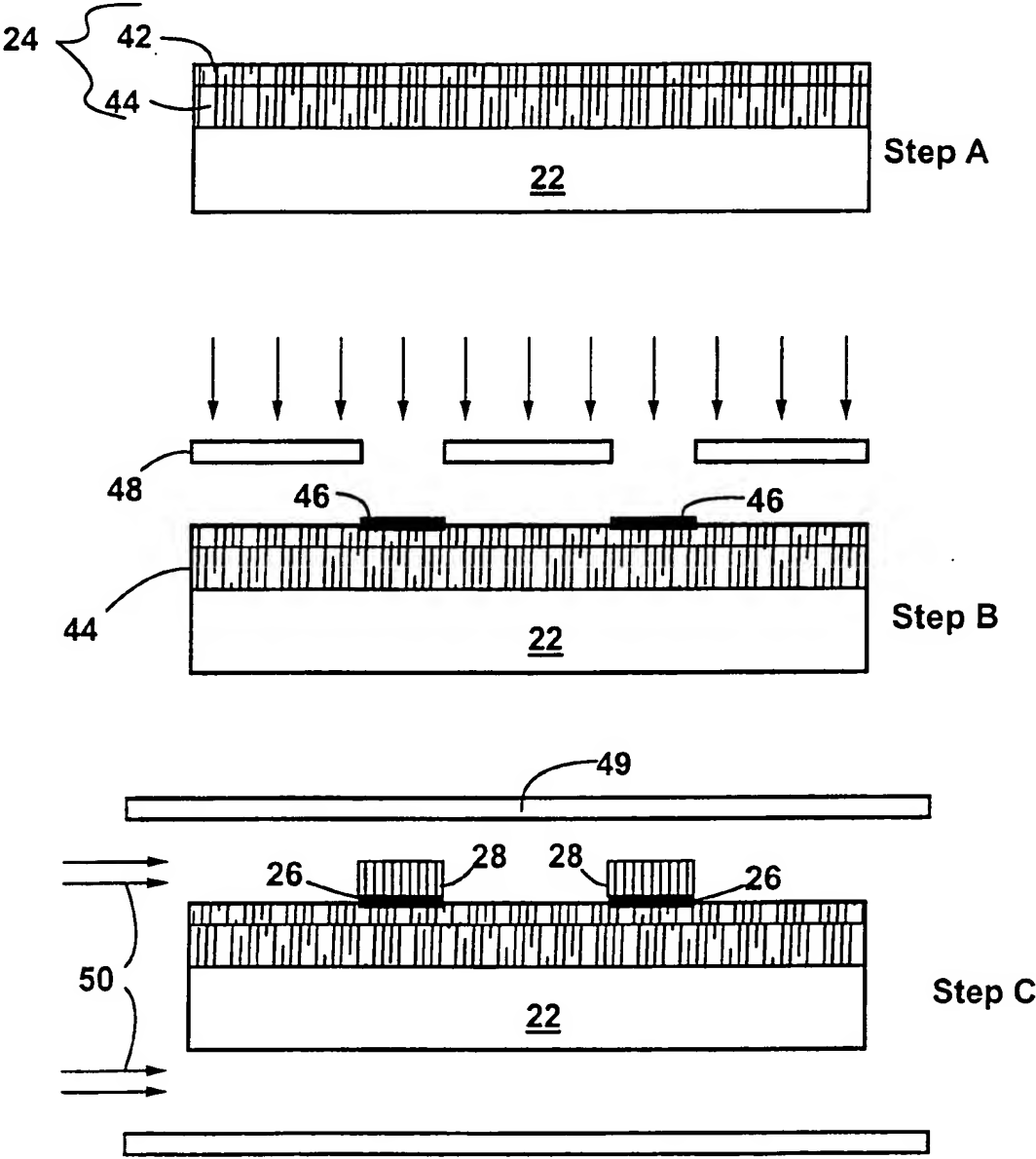


Fig. 3

3/3

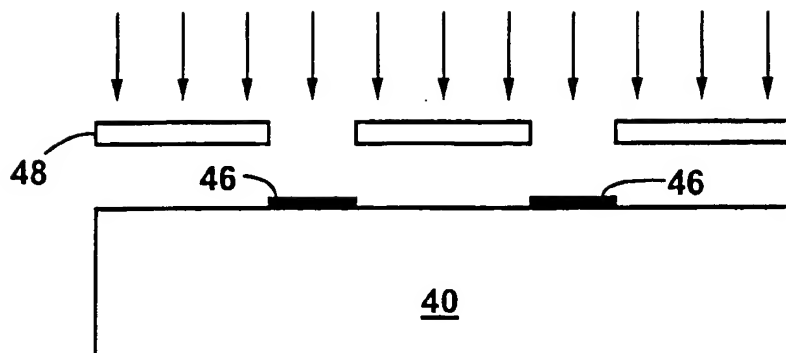


Fig. 4

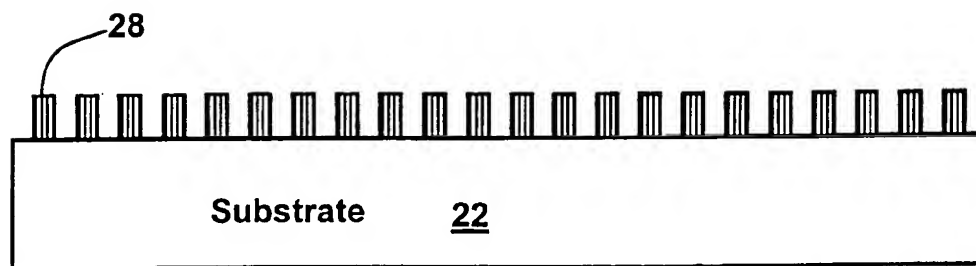


Fig. 5

INTERNATIONAL SEARCH REPORT

 International application No.
 PCT/US99/26232

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : H01J 01/30; C23C 16/00

US CL : Please See Extra Sheet.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 313/309, 310, 336, 351, 346R, 494; 427/ 58, 255.6, 508, 521, 577, 578, 579, 249, 250, 255, 255.1; 204/192.1, 192.38

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONEElectronic data base consulted during the international search (name of data base and, where practicable, search terms used)
NONE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X, P	US 5,872,422 A (XU et al.) 16 February 1999 (16.02.99), col. 5, line 8-col. 21, line 67.	1-24
X, P	US 5,973,444 A (XU et al.) 26 October 1999 (26.10.99), col. 5, line 9-col. 22, line 6.	1-24
A	US 4,272,699 A (FAUBEL et al.) 09 June 1981 (09.06.81), See entire document.	1-24
A	US 5,697,827 A (RABINOWITZ) 16 December 1997 (16.12.97), See entire document.	1-24
A	US 5,726,524 A (DEBE) 10 March 1998 (10.03.98), See entire document.	1-24
A	US 5,773,921 A (KEESMANN et al.) 30 June 1998 (30.06.98), See entire document.	1-24

☐ Further documents are listed in the continuation of Part C.
 ☐ See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Z" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

15 JANUARY 2000

Date of mailing of the international search report

17 FEB 2000

 Name and mailing address of the ISA/US
 Commissioner of Patents and Trademarks
 Box PCT
 Washington, D.C. 20231

Facsimile No. (703) 305-3230

Authorized officer

MACK HAYNES

Telephone No. (703) 308-0956

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US99/26232

A. CLASSIFICATION OF SUBJECT MATTER:

US CL :

313/309, 310, 336, 351, 346R, 494; 427/ 58, 255.6, 508, 521, 577, 578, 579, 249, 250, 255, 255.1; 204/192.1,
192.38